

PATENT

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Title: CATALYST FOR  
POLYMERIZATION AND  
CO-POLYMERIZATION  
OF ETHYLENE

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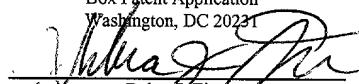
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PRELIMINARY AMENDMENT

Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir:

Please amend the above-captioned application as follows:

*In the Specification:*

Please replace pages 1-19 of the specification with pages 1-20 of the enclosed substitute sheets. Applicant has also submitted herewith a strikethrough version of the specification indicating the amendments.

***In the Claims:***

**Claims**

What is claimed is:

Please amend the claims as follows:

1. A catalyst for polymerization and co-polymerization of ethylene, wherein said catalyst is produced by [means of] the method comprising:
  - (i) preparing a magnesium solution by contact-reacting a halogenated magnesium compound with alcohol;
  - (ii) reacting said solution with an ester compound having at least one hydroxy group, or a phosphorus compound and a silicon compound having alkoxy groups, and then producing a solid component by adding a mixture of a titanium compound and a silicon compound thereto; and
  - (iii) reacting said solid component with an aluminum compound, and then reacting the same with a titanium compound, or a titanium compound and a vanadium compound.
2. The catalyst [for polymerization and co-polymerization of ethylene according to] of Claim 1, wherein [said] the ester compound containing at least one hydroxy group [is] comprises an unsaturated aliphatic acid ester having at least one hydroxy group, [which is selected from the group consisting of 2-hydroxy ethylacrylate, 2-hydroxy ethylmethacrylate, 2-hydroxy propylacrylate, 2-hydroxy propylmethacrylate, 4-hydroxy butylacrylate, pentaerithritol tri-acrylate;] an aliphatic monoester or polyester having at least one hydroxy group, [which is selected from the group consisting of 2-hydroxy ethyl acetate, methyl 3-hydroxy butylate, ethyl

3-hydroxy butylate, methyl 2-hydroxy isobutylate, ethyl 2-hydroxy isobutylate, methyl-3-hydroxy-2-methyl propionate, 2,2-dimethyl-3-hydroxy propionate, ethyl-6-hydroxy hexanoate, t-butyl-2-hydroxy isobutylate, diethyl-3-hydroxy glutarate, ethyl-lactate, isopropyl lactate, butyl-isobutyl lactate, isobutyl lactate, ethyl mandelate, dimethyl ethyl tartrate, ethyl tartrate, dibutyl tartrate, diethyl citrate, triethyl citrate, ethyl-2-hydroxy-caproate, diethyl *bis*-(hydroxymethyl) malonate;] an aromatic ester having at least one hydroxy group, [which is selected from the group consisting of 2-hydroxy ethyl benzoate, 2-hydroxy ethyl salicylate, methyl-4-(hydroxy methyl) benzoate, methyl-4-hydroxy benzoate, ethyl-3-hydroxy benzoate, 4-methyl salicylate, ethyl salicylate, phenyl salicylate, propyl-4-hydroxy benzoate, phenyl-3-hydroxy naphthanoate, monoethylene glycol monobenzoate, diethylene glycol monobenzoate, triethylene glycol monobenzoate;] or an alicyclic ester having at least one hydroxy group [as in hydroxy butyl-lactone;

wherein said phosphorus compound is a compound expressed by  $PX_aR^1_b(OR^2)_c$ , or  $POX_dR^3_e(OR^4)_f$ , where X is a halogen atom; and  $R^1$ ,  $R^2$ ,  $R^3$  or  $R^4$  is a hydrocarbon of an alkyl, or alkenyl or aryl group, having 1 ~ 20 carbon atoms, each of which can be same or different from one another, with  $a + b + c = 3$ ,  $0 \leq a \leq 3$ ,  $0 \leq b \leq 3$ ,  $0 \leq c \leq 3$ ,  $d + e + f = 3$ ,  $0 \leq d \leq 3$ ,  $0 \leq e \leq 3$ , and  $0 \leq f \leq 3$ ; and

wherein said silicon compound having alkoxy groups is a compound of a general formula of  $R_nSi(OR)_{4-n}$ , where R is a hydrocarbon group having 1~12 carbon atoms, and n is a natural number of 1~3].

3. The catalyst [for polymerization and co-polymerization of ethylene according to Claim 2] of claim 1, wherein [said] the phosphorus compound [is selected from the group consisting of] comprises phosphorus trichloride, phosphorus tribromide, diethylchlorophosphite, diphenylchlorophosphite, diethylbromophosphite, diphenylbromophospite, dimethylchlorophosphite, phenylchlorophosphite, trimethylphosphite, triethylphosphite, tri-n-

butylphosphite, trioctylphosphite, tridecylphosphite, triphenylphosphite, triethylphosphate, tri-n-butylphosphate, [and] or triphenylphosphate.

4. The catalyst [for polymerization and co-polymerization of ethylene according to Claim 2] of claim 1, wherein [said] the silicone compound having alkoxy groups [is selected from the group consisting of] comprises dimethyldimethoxysilane, dimethyldiethoxysilane, diphenyldimethoxysilane, methylphenylmethoxysilane, diphenyldiethoxysilane, ethyltrimethoxysilane, vinyltrimethoxysilane, methyltrimethoxysilane, phenyltrimethoxysilane, methyltriethoxysilane, ethyltriethoxysilane, vinyltriethoxysilane, butyltriethoxysilane, phenyltriethoxysilane, ethyltriisopropoxysilane, vinyltributoxysilane, ethylsilicate, butylsilicate, or methyltriaryloxy silane.

5. The catalyst [for polymerization and co-polymerization of ethylene according to] of Claim 1, wherein [said] the titanium compound is represented by a general formula of  $Ti(OR)_aX_4-a$ , where R is a hydrocarbon group, X for a halogen atom, and a for a natural number of  $0 \leq a \leq 4$ ; and wherein said silicon compound is represented by a general formula of  $R_nSiCl_{n-4}$ , where R is hydrogen, an aryl, alkoxy, haloalkyl or alkyl group having 1~10 carbon atoms, or a halosilylalkyl or halosilyl group having 1~8 carbon atoms, and n for a natural number of  $0 \leq n \leq 4$ .

6. The catalyst [for polymerization and co-polymerization of ethylene according to Claim 5] of claim 1, wherein [said] the titanium compound [is] comprises a 4-halogenated titanium, [which is selected from the group consisting of  $TiCl_4$ ,  $TiBr_4$ , and  $TiI_4$ ; ] a 3-halogenated alkoxytitanium, [which is selected from the group consisting of  $Ti(OCH_3)Cl_3$ ,  $Ti(OC_2H_5)Cl_3$ ,  $Ti(OC_2H_5)Br_3$ , and  $Ti(O(i-C_4H_9))Br_3$ ; ] a 2-halogenated alkoxytitanium, [which is selected from the group consisting of  $Ti(OCH_3)_2Cl_2$ ,  $Ti(OC_2H_5)_2Cl_2$ ,  $Ti(O(i-C_4H_9))_2Cl_2$ , and  $Ti(OC_2H_5)_2Br_2$ ; and] or a tetralkoxy titanium, [which is selected from the group consisting of  $Ti(OCH_3)_4$ ,  $Ti(OC_2H_5)_4$ , and  $Ti(OC_4H_9)_4$ ;] and wherein [said] the silicon compound is silicon tetrachloride, [or] a trichlorosilane, [which is selected from the group consisting of methyltrichlorosilane,

ethyltrichlorosilane, and phenyl-trichlorosilane;] a dichlorosilane, [which is selected from the group consisting of dimethylchlorosilane, diethyldichlorosilane, diphenyldichlorosilane, and methylphenyldichlorosilane;] or a monochlorosilane [as in trimethylchlorosilane].

7. The catalyst [for polymerization and co-polymerization of ethylene according to Claim 6] of claim 1, wherein [said] the titanium compound is titanium tetrachloride, and [said] the silicon compound is silicon tetrachloride.

8. The catalyst [for polymerization and co-polymerization of ethylene according to] of Claim 1, wherein the amount of the mixture of said titanium compound and said silicon compound is 0.1 ~ 200 mol per mole of said halogenated magnesium compound, and the molar ratio of said titanium compound to said silicon compound in the mixture is 0.05 ~ 0.95.

9. The catalyst [for polymerization and co-polymerization of ethylene according to] of Claim 1, wherein [said] the aluminum compound [is] comprises a trialkylaluminum having an alkyl group of 1~6 carbon atoms, [which is selected from the group consisting of triethylaluminum and triisobutylaluminum;] an aluminum compound having one more halogen, [which is selected from the group consisting of ethylaluminum dichloride, diehtylaluminum chloride, and ethylaluminum sesquichloride;] or [the mixture] mixtures thereof.

10. The catalyst [for polymerization and co-polymerization of ethylene according to] of Claim 1, wherein said vanadium compound is a compound with the maximum atomic valence of 4, or the maximum atomic valence of VO of a vanadyl group of 3, having a general formula of  $V(OR^4)_{4-n}X_n$ , or  $VO(OR^4)_{3-m}X_m$ , where  $R^4$  is an aliphatic or aromatic hydrocarbon group having 1~14 carbons, or  $COR^5$ , where  $R^5$  is an aliphatic or aromatic hydrocarbon group having 1~14 carbons, wherein X is C1, Br or I; n is an integer of 0~4, or the ratio thereof; and m is an integer of 0~3, or the ratio thereof.

Please add the following claims:

11. (New) The catalyst of claim 1, wherein the ester compound containing at least one hydroxy group comprises 2-hydroxy ethylacrylate, 2-hydroxy ethylmethacrylate, 2-hydroxy propylacrylate, 2-hydroxy propylmethacrylate, 4-hydroxy butylacrylate, pentaerithritol triacrylate, 2-hydroxy ethyl acetate, methyl 3-hydroxy butylate, ethyl 3-hydroxy butylate, methyl 2-hydroxy isobutylate, ethyl 2-hydroxy isobutylate, methyl-3-hydroxy-2-methyl propionate, 2,2-dimethyl-3-hydroxy propionate, ethyl-6-hydroxy hexanoate, t-butyl-2-hydroxy isobutylate, diethyl-3-hydroxy glutarate, ethyl-lactate, isopropyl lactate, butyl-isobutyl lactate, isobutyl lactate, ethyl mandelate, dimethyl ethyl tartrate, ethyl tartrate, dibutyl tartrate, diethyl citrate, triethyl citrate, ethyl-2-hydroxy-caproate, diethyl bis-(hydroxymethyl) malonate, 2-hydroxy ethyl benzoate, 2-hydroxy ethyl salicylate, methyl-4-(hydroxy methyl) benzoate, methyl-4-hydroxy benzoate, ethyl-3-hydroxy benzoate, 4-methyl salicylate, ethyl salicylate, phenyl salicylate, propyl-4-hydroxy benzoate, phenyl-3-hydroxy naphthanoate, monoethylene glycol monobenzoate, diethylene glycol monobenzoate, triethylene glycol monobenzoate, or hydroxy butyl-lactone.

12. (New) The catalyst of claim 1, wherein the phosphorus compound is a compound expressed by  $PX_aR^1_b(OR^2)_c$ , or  $POX_dR^3_e(OR^4)_f$ , where X is a halogen atom; and R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup> or R<sup>4</sup> is a hydrocarbon of an alkyl, or alkenyl or aryl group, having 1 ~ 20 carbon atoms, each of which can be same or different from one another, with  $a + b + c = 3$ ,  $0 \leq a \leq 3$ ,  $0 \leq b \leq 3$ ,  $0 \leq c \leq 3$ ,  $d + e + f = 3$ ,  $0 \leq d \leq 3$ ,  $0 \leq e \leq 3$ , and  $0 \leq f \leq 3$ .

13. (New) The catalyst of claim 1, wherein the silicon compound having alkoxy groups is a compound of a general formula of  $R_nSi(OR)_{4-n}$ , where R is a hydrocarbon group having 1~12 carbon atoms, and n is a natural number of 1~3].

14. (New) The catalyst of claim 1, wherein the titanium compound comprises  $TiCl_4$ ,  $TiBr_4$ , and  $TiI_4$ ,  $Ti(OCH_3)Cl_3$ ,  $Ti(OC_2H_5)Cl_3$ ,  $Ti(OC_2H_5)Br_3$ ,  $Ti(O(i-C_4H_9))Br_3$ ,  $Ti(OCH_3)_2Cl_2$ ,  $Ti(OC_2H_5)_2Cl_2$ ,  $Ti(O(i-C_4H_9))_2Cl_2$ ,  $Ti(OC_2H_5)_2Br_2$ ,  $Ti(OCH_3)_4$ ,  $Ti(OC_2H_5)_4$ , or  $Ti(OC_4H_9)_4$ .

15. (New) The catalyst of claim 1, wherein the silicon compound comprises silicon tetrachloride, methyltrichlorosilane, ethyltrichlorosilane, phenyl-trichlorosilane, dimethylchlorosilane, diethyldichlorosilane, diphenyldichlorosilane, methylphenyldichlorosilane, or trimethylchlorosilane.

16. (New) The catalyst of claim 1, wherein the aluminum compound comprises triethylaluminum, triisobutylaluminum, ethylaluminum dichloride, diethylaluminum chloride, ethylaluminum sesquichloride, or mixtures thereof.

17. (New) A method of forming a catalyst for polymerization and co-polymerization of ethylene comprising:

preparing a magnesium solution by contact-reacting a halogenated magnesium compound with alcohol;

reacting said solution with an ester compound having at least one hydroxy group, or a phosphorus compound and a silicon compound having alkoxy groups, and then producing a solid component by adding a mixture of a titanium compound and a silicon compound thereto; and

reacting said solid component with an aluminum compound, and then reacting the same with a titanium compound, or a titanium compound and a vanadium compound.

18. (New) The method of claim 17, wherein the ester compound containing at least one hydroxy group comprises an unsaturated aliphatic acid ester having at least one hydroxy group, an aliphatic monoester or polyester having at least one hydroxy group, an aromatic ester having at least one hydroxy group, or an alicyclic ester having at least one hydroxy group.
19. (New) The method of claim 17, wherein the phosphorus compound comprises phosphorus trichloride, phosphorus tribromide, diethylchlorophosphite, diphenylchlorophosphite, diethylbromophosphite, diphenylbromophosphite, dimethylchlorophosphite, phenylchlorophosphite, trimethylphosphite, triethylphosphite, tri-n-butylphosphite, trioctylphosphite, tridecylphosphite, triphenylphosphite, triethylphosphate, tri-n-butylphosphate, or triphenylphosphate.
20. (New) The method of claim 17, wherein the silicone compound having alkoxy groups comprises dimethyldimethoxysilane, dimethyldiethoxysilane, diphenyldimethoxysilane, methylphenylmethoxysilane, diphenyldiethoxysilane, ethyltrimethoxysilane, vinyltrimethoxysilane, methyltrimethoxysilane, phenyltrimethoxysilane, methyltriethoxysilane, ethyltriethoxysilane, vinyltriethoxysilane, butyltriethoxysilane, phenyltriethoxysilane, ethyltriisopropoxysilane, vinyltributoxysilane, ethylsilicate, butylsilicate, or methyltriaryloxylosilane.
21. (New) The method of claim 17, wherein the titanium compound is represented by a general formula of  $Ti(OR)_aX_{4-a}$ , where R is a hydrocarbon group, X for a halogen atom, and a for a natural number of  $0 \leq a \leq 4$ ; and wherein the silicon compound is represented by a general formula of  $R_nSiCl_{n-4}$ , where R is hydrogen, an aryl, alkoxy, haloalkyl or alkyl group having 1~10 carbon atoms, or a halosilylalkyl or halosilyl group having 1~8 carbon atoms, and n for a natural number of  $0 \leq n \leq 4$ .

22. (New) The method of claim 17, wherein the titanium compound comprises a 4-halogenated titanium, a 3-halogenated alkoxytitanium, a 2-halogenated alkoxytitanium, or a tetralkoxy titanium, and wherein the silicon compound is silicon tetrachloride, a trichlorosilane, a dichlorosilane, or a monochlorosilane.
23. (New) The method of claim 17, wherein said titanium compound is titanium tetrachloride, and said silicon compound is silicon tetrachloride.
24. (New) The method of claim 17, wherein the amount of the mixture of said titanium compound and said silicon compound is 0.1 ~ 200 mol per mole of said halogenated magnesium compound, and the molar ratio of said titanium compound to said silicon compound in the mixture is 0.05 ~ 0.95.
25. (New) The method of claim 17, wherein the aluminum compound comprises a trialkylaluminum having an alkyl group of 1~6 carbon atoms, an aluminum compound having one or more halogen atoms, or mixtures thereof.
26. (New) The method of claim 17, wherein the vanadium compound is a compound with a maximum atomic valence of 4, or a maximum atomic valence of VO of a vanadyl group of 3, having a general formula of  $V(OR^4)_{4-n}X_n$ , or  $VO(OR^4)_{3-m}X_m$ , where  $R^4$  is an aliphatic or aromatic hydrocarbon group having 1~14 carbons, or  $COR^5$ , where  $R^5$  is an aliphatic or aromatic hydrocarbon group having 1~14 carbons, wherein X is Cl, Br or I; n is an integer of 0~4, or the ratio thereof; and m is an integer of 0~3, or the ratio thereof.
27. (New) The method of claim 17, wherein the ester compound containing at least one hydroxy group comprises 2-hydroxy ethylacrylate, 2-hydroxy ethylmethacrylate, 2-hydroxy propylacrylate, 2-hydroxy propylmethacrylate, 4-hydroxy butylacrylate, pentaerithritol tri-acrylate, 2-hydroxy ethyl acetate, methyl 3-hydroxy butylate, ethyl 3-hydroxy butylate, methyl 2-

hydroxy isobutylate, ethyl 2-hydroxy isobutylate, methyl-3-hydroxy-2-methyl propionate, 2,2-dimethyl-3-hydroxy propionate, ethyl-6-hydroxy hexanoate, t-butyl-2-hydroxy isobutylate, diethyl-3-hydroxy glutarate, ethyl-lactate, isopropyl lactate, butyl-isobutyl lactate, isobutyl lactate, ethyl mandelate, dimethyl ethyl tartrate, ethyl tartrate, dibutyl tartrate, diethyl citrate, triethyl citrate, ethyl-2-hydroxy-caproate, diethyl *bis*-(hydroxymethyl) malonate, 2-hydroxy ethyl benzoate, 2-hydroxy ethyl salicylate, methyl-4-(hydroxy methyl) benzoate, methyl-4-hydroxy benzoate, ethyl-3-hydroxy benzoate, 4-methyl salicylate, ethyl salicylate, phenyl salicylate, propyl-4-hydroxy benzoate, phenyl-3-hydroxy naphthanoate, monoethylene glycol monobenzoate, diethylene glycol monobenzoate, triethylene glycol monobenzoate, or hydroxy butyl-lactone.

28. (New) The method of claim 17, wherein the phosphorus compound is a compound expressed by  $PX_aR^1_b(OR^2)_c$ , or  $POX_dR^3_e(OR^4)_f$ , where X is a halogen atom; and R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup> or R<sup>4</sup> is a hydrocarbon of an alkyl, or alkenyl or aryl group, having 1 ~ 20 carbon atoms, each of which can be same or different from one another, with  $a + b + c = 3$ ,  $0 \leq a \leq 3$ ,  $0 \leq b \leq 3$ ,  $0 \leq c \leq 3$ ,  $d + e + f = 3$ ,  $0 \leq d \leq 3$ ,  $0 \leq e \leq 3$ , and  $0 \leq f \leq 3$ .

29. (New) The method of claim 17, wherein the silicon compound having alkoxy groups is a compound of a general formula of  $R_nSi(OR)_{4-n}$ , where R is a hydrocarbon group having 1~12 carbon atoms, and n is a natural number of 1~3].

30. (New) The method of claim 17, wherein the titanium compound comprises  $TiCl_4$ ,  $TiBr_4$ , and  $TiI_4$ ,  $Ti(OCH_3)Cl_3$ ,  $Ti(OC_2H_5)Cl_3$ ,  $Ti(OC_2H_5)Br_3$ ,  $Ti(O(i-C_4H_9))Br_3$ ,  $Ti(OCH_3)_2Cl_2$ ,  $Ti(OC_2H_5)_2Cl_2$ ,  $Ti(O(i-C_4H_9))_2Cl_2$ ,  $Ti(OC_2H_5)_2Br_2$ ,  $Ti(OCH_3)_4$ ,  $Ti(OC_2H_5)_4$ , or  $Ti(OC_4H_9)_4$ .

31. (New) The method of claim 17, wherein the silicon compound comprises silicon tetrachloride, methyltrichlorosilane, ethyltrichlorosilane, phenyl-trichlorosilane,

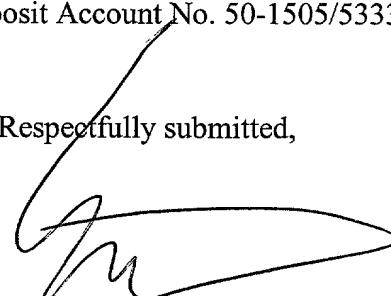
dimethylchlorosilane, diethyldichlorosilane, diphenyldichlorosilane, methylphenyldichlorosilane, or trimethylchlorosilane.

32. (New) The method of claim 17, wherein the aluminum compound comprises triethylaluminum, triisobutylaluminum, ethylaluminum dichloride, diethylaluminum chloride, ethylaluminum sesquichloride, or mixtures thereof.

Please replace pages 20-23 of the claims with pages 21-28 of the enclosed substitute sheets.

It is believed that no fees are due in connection with the filing of this Preliminary Amendment. However, if any fees are due, the Assistant Commissioner is hereby authorized to deduct said fees from Conley, Rose & Tayon Deposit Account No. 50-1505/5333-01300/EBM.

Respectfully submitted,



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